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EXAMINER

OH, TAYLOR V

ART UNIT PAPER NUMBER

1625

DATE MAILED: 03/21/2006

Please find below and/or attached an Office communication concerning this application or proceeding.

**Office Action Summary**

Application No.

10/621,399

Applicant(s)

YONEHARA ET AL.

Examiner

Taylor Victor Oh

Art Unit

1625

-- The MAILING DATE of this communication appears on the cover sheet with the correspondence address --

**Period for Reply**

A SHORTENED STATUTORY PERIOD FOR REPLY IS SET TO EXPIRE 3 MONTH(S) OR THIRTY (30) DAYS, WHICHEVER IS LONGER, FROM THE MAILING DATE OF THIS COMMUNICATION.

- Extensions of time may be available under the provisions of 37 CFR 1.136(a). In no event, however, may a reply be timely filed after SIX (6) MONTHS from the mailing date of this communication.
- If NO period for reply is specified above, the maximum statutory period will apply and will expire SIX (6) MONTHS from the mailing date of this communication.
- Failure to reply within the set or extended period for reply will, by statute, cause the application to become ABANDONED (35 U.S.C. § 133). Any reply received by the Office later than three months after the mailing date of this communication, even if timely filed, may reduce any earned patent term adjustment. See 37 CFR 1.704(b).

**Status**

- 1) ☒ Responsive to communication(s) filed on 21 December 2005.
- 2a) ☐ This action is **FINAL**. 2b) ☒ This action is non-final.
- 3) ☐ Since this application is in condition for allowance except for formal matters, prosecution as to the merits is closed in accordance with the practice under *Ex parte Quayle*, 1935 C.D. 11, 453 O.G. 213.

**Disposition of Claims**

- 4) ☒ Claim(s) 1-20 is/are pending in the application.
- 4a) Of the above claim(s) 11-15 is/are withdrawn from consideration.
- 5) ☐ Claim(s) \_\_\_\_\_ is/are allowed.
- 6) ☒ Claim(s) 1-10 and 16-20 is/are rejected.
- 7) ☐ Claim(s) \_\_\_\_\_ is/are objected to.
- 8) ☐ Claim(s) \_\_\_\_\_ are subject to restriction and/or election requirement.

**Application Papers**

- 9) ☐ The specification is objected to by the Examiner.
- 10) ☐ The drawing(s) filed on \_\_\_\_\_ is/are: a) ☐ accepted or b) ☐ objected to by the Examiner.  
Applicant may not request that any objection to the drawing(s) be held in abeyance. See 37 CFR 1.85(a).  
Replacement drawing sheet(s) including the correction is required if the drawing(s) is objected to. See 37 CFR 1.121(d).
- 11) ☐ The oath or declaration is objected to by the Examiner. Note the attached Office Action or form PTO-152.

**Priority under 35 U.S.C. § 119**

- 12) ☒ Acknowledgment is made of a claim for foreign priority under 35 U.S.C. § 119(a)-(d) or (f).
- a) ☒ All b) ☐ Some \* c) ☐ None of:
1. ☒ Certified copies of the priority documents have been received.
  2. ☐ Certified copies of the priority documents have been received in Application No. \_\_\_\_\_.
  3. ☐ Copies of the certified copies of the priority documents have been received in this National Stage application from the International Bureau (PCT Rule 17.2(a)).

\* See the attached detailed Office action for a list of the certified copies not received.

**Attachment(s)**

- |  |   |
|--|---|
| 1) <input type="checkbox"/> Notice of References Cited (PTO-892)   | 4) <input type="checkbox"/> Interview Summary (PTO-413)<br>Paper No(s)/Mail Date. _____ |
| 2) <input type="checkbox"/> Notice of Draftsperson's Patent Drawing Review (PTO-948)   | 5) <input type="checkbox"/> Notice of Informal Patent Application (PTO-152)             |
| 3) <input checked="" type="checkbox"/> Information Disclosure Statement(s) (PTO-1449 or PTO/SB/08)<br>Paper No(s)/Mail Date <u>8/05, 10/03, 1/04</u> . | 6) <input type="checkbox"/> Other: _____  |

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**The Status of Claims :**

Claims 1-20 are pending.

Claims 1-10 and 16-20 have been rejected.

Claims 11-15 have been withdrawn from consideration.

**DETAILED ACTION**

1. Claims 1-10 and 16-20 are under consideration in this Office Action.

Priority

2. It is noted that the foreign priority document , Japan 2002-209987 is in the file.

Drawings

3. None.

***Election/Restrictions***

Applicant's election without traverse of Group I (claims 1-10 and 16-20) on 12/21/05 is acknowledged.

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Claims 11-15 (Group II) are withdrawn from further consideration pursuant to 37 CFR 1.142(b) as being drawn to a nonelected group II, there being no allowable generic or linking claim.

***Claim Rejections - 35 USC § 112***

***Claim Rejections - 35 USC § 112***

The following is a quotation of the second paragraph of 35 U.S.C. 112:

The specification shall conclude with one or more claims particularly pointing out and distinctly claiming the subject matter which the applicant regards as his invention.

Claims 9 and 20 are rejected under 35 U.S.C. 112, second paragraph, as being indefinite for failing to particularly point out and distinctly claim the subject matter which applicant regards as the invention.

In claims 9 and 20 , the phrase “ tungsten atom-containing heteropolyoxometallate anions” is recited. The term “containing” is vague and indefinite because the term “containingwould mean that there are other components besides tungsten atom; there is uncertainty as to what other compounds are present in the product. Furthermore, It is well-settled that the term “ containing ” do not exclude the presence of other ingredients than the one recited. Ex parte Muench , 79 USPQ 92 (PTO Bd. App. 1948). Therefore, an appropriate correction is required.

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***Claim Rejections - 35 USC § 102***

The following is a quotation of the appropriate paragraphs of 35 U.S.C. 102 that form the basis for the rejections under this section made in this Office action:

A person shall be entitled to a patent unless –

(b) the invention was patented or described in a printed publication in this or a foreign country or in public use or on sale in this country, more than one year prior to the date of application for patent in the United States.

1. Claims 1-10 , and 17-20 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Watanabe et al (J. of Molecular Catalysis A : Chem.. 145 (1999), p. 281-289).

Watanabe et al discloses the followings (see an abstract page 281):

The epoxidation of alkenes with hydrogen peroxide or molecular oxygen, in the presence of aldehyde catalyzed by heteropolyoxometalate (HPOM) salts and layered double hydroxides (LDHs) containing HPOM anions as pillars, has been investigated. In the case of oxidation with hydrogen peroxide, a lacunary Keggin-ion,  $\text{SiW}_{11}\text{O}_{39}^{8-}$ , was more active than  $\text{SiW}_{12}\text{O}_{40}^{4-}$ . For this reaction system using hydrogen peroxide, intercalated  $\text{SiW}_{11}\text{O}_{39}^{8-}$  showed higher stability in structure and activity than free  $\text{K}_8\text{SiW}_{11}\text{O}_{39}$ . For the oxidation using molecular oxygen and aldehyde,  $\text{SiW}_{12}\text{O}_{40}^{4-}$  showed higher activity than  $\text{SiW}_{11}\text{O}_{39}^{8-}$ . It was confirmed that  $\text{SiW}_{12}\text{O}_{40}^{4-}$  was intercalated between the brucite-type layers of the LDHs without change in structure. Ni-containing LDHs without any interstitial polyoxometalate showed high activity for the oxidation of cyclohexene with the combination of  $\text{O}_2$  and aldehyde. © 1999 Elsevier Science B.V. All rights reserved.

### 2.3.1. Oxidation of alkenes with $H_2O_2$

Oxidation reactions of alkenes were carried out in a 100-ml flask containing catalyst (1 mmol of polymetal), alkene (100 ml) and 10 wt.%  $H_2O_2$  (100 mmol) in tributyl phosphate (TBP), at 343 K for 3 h. A non-aqueous  $H_2O_2$ -TBP solution was prepared by adding 30%  $H_2O_2$  aqueous solution to TBP, followed by evaporation of water under reduced pressure at 323 K [16].

Table 1  
Chemical compositions and methods of synthesis

Sample	Chemical composition (molar ratio) $M^{2+}:Al^{3+}:metal^a$
$Zn_2Al-SiW_{12}O_{40}$	2.0:1.0:0.44
$Zn_3Al-SiW_{12}O_{40}$	3.0:1.0:0.78
$Zn_4Al-SiW_{12}O_{40}$	3.3:1.0:0.12
$Zn_3Al-SiW_{11}O_{39}$	2.9:1.0:1.4
$Zn_2Al-SiW_9O_{34}$	2.0:1.0:1.2
$Mg_2Al-SiW_{12}O_{40}$	2.3:1.0:1.2
$Ni_2Al-SiW_{12}O_{40}$	1.3:1.0:1.6
$Ni_2Al-TA^b$	2.2:1.0:0

<sup>a</sup>Polymetal in HPOM.

<sup>b</sup>TA = terephthalate dianion.

(see page 283, table 1 , lower left col. ). This is identical with the claims.

2. Claims 1-7, 9-10 ,17-18, and 20 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Sakamoto et al (Tetrahedron Letters 41 (2000), p.10009-10012).

Sakamoto et al discloses the followings :

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It was found that the catalytic epoxidation of various olefins in 15% aqueous  $H_2O_2$  using the solid catalyst does occur in various efficiencies and selectivities depending on different modifications of silica gel as well as on the organic counter cations of tungstophosphate. Table 1 summarises the catalytic epoxidation of 1-octene in 15% aqueous  $H_2O_2$  using the various catalysts prepared (Cat. 1-13). As a typical run, a mixture of 1-octene (3 mmol), Cat. 1 (0.5 mol%) and 15%  $H_2O_2$  (6 mmol) was heated at  $90^\circ C$  for 10 h. GLC analysis showed the quantitative formation of 1,2-epoxyoctane at the complete consumption of 1-octene. Prominent features are as follows: (1) Modification of silica gel with both  $Ph_3SiOEt$  and

(see page 10010, a lower section).

Table 1  
Epoxidation of 1-octene with 15% aqueous  $H_2O_2$ <sup>a</sup>

Cat.	Catalyst <sup>b</sup>			Conversion of 1-octene (%) <sup>c</sup>	Selectivity of epoxide (%) <sup>c</sup>
	Polyoxometalate $[R]_x(PW_{12}O_{40})^{x-}$	Silane coupling agent (SA)	Alkylating agent (AA) $(CH_3)_2NCH(OX)_2$		
1	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = benzyl	100	> 98
2	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	None	59	97
3	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	None	X = benzyl	45	97
4	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	None	None	18	96
5 <sup>d</sup>	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = benzyl	72	> 98
6 <sup>e</sup>	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = benzyl	47	> 98
7	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	$Ph_2Si(OC_2H_5)_2$	X = benzyl	79	> 98
8	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = n-butyl	81	97
9	$R = \pi-C_3H_5N^+(CH_2)_{15}CH_3$	$Ph_3SiOC_2H_5$	X = cyclohexyl	78	97
10	$R = C_{12}H_{25}(CH_3)_3N^+$	$Ph_3SiOC_2H_5$	X = benzyl	90	97
11	$R = CH_3(C_8H_{17})_3N^+$	$Ph_3SiOC_2H_5$	X = benzyl	88	97
12	$R = (C_4H_9)_4N^+$	$Ph_3SiOC_2H_5$	X = benzyl	41	97
13	R = H	$Ph_3SiOC_2H_5$	X = benzyl	21	96

<sup>a</sup> For reactions of 3 mmol 1-octene, 0.015 mmol of catalyst (0.5 mol%) and 6 mmol 15%  $H_2O_2$  at  $90^\circ C$  for 10 h.

<sup>b</sup> Unless otherwise stated, SA:AA = 1:1.

<sup>c</sup> By GC analysis using  $Bu_2O$  as internal standard.

<sup>d</sup> SA:AA = 3:1.

<sup>e</sup> SA:AA = 1:3.

(see page 10011, table 1). This is identical with the claims.

3. Claims 1-5, 7-10, and 18-20 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Briot et al (J. Mater. Chem. 2000, 10, p. 953-958).

Briot et al discloses the followings (see page 957) :

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The results of cyclooctene epoxidation over W-containing MCM-41 are listed in Table 2. Blank experiments with purely siliceous MCM-41 show that it is virtually inactive for this reaction at room temperature. In contrast, W-containing MCM-41 catalysts are very active, even at room temperature. Comparison of A1c and Bc indicates that materials synthesized by route A are distinctly more active. This can be explained by the presence of less active  $\text{WO}_3$  material in the Bc sample. More interesting is the result obtained with sample A2c. Despite the high tungsten loading in A2c ( $(\text{Si}:\text{W})_{\text{exp}} = 20:1$ ) and the large amount of tungsten used during the catalysis test ( $(\text{Substrate}:\text{W}) = 42:1$ ), cyclooctene conversion is less than that observed with A1c. This result can be understood by considering that sample A2c does not only contain isolated W species, but also tungsten oxide and/or HPA species which are poorer precatalysts for this reaction. Catalysis results match those obtained by different physicochemical techniques, especially Raman spectroscopy. Furthermore, atomic emission spectroscopic analysis of the epoxidation solutions indicates that there is reduced leaching (quantitative analysis is in progress) of W into the solutions. Work is in progress to obtain materials resistant to nucleophilic attack and to run tests with weak nucleophiles.

Table 1 Tungsten content, (Si/W) molar ratio, unit cell parameters, specific surface area, mean pore diameter ( $\phi_{\text{BHH}}$ ) and wall thickness ( $w$ )

Run	Route	(Si:W) <sub>inhib</sub>	Si (wt.%)	W (wt.%)	(Si:W) <sub>exp</sub>	$a_0/\text{\AA}$	$S_{\text{DET}}/\text{m}^2 \text{g}^{-1}$	$\phi_{\text{BHH}}/\text{\AA}$	$w/\text{\AA}$
All Si									
A1	oxo peroxo	40	39.4	8.2	31	45.5 (36)	1384 ± 45	16	19.5
A2	oxo peroxo	20	36.2	12.5	19	46 (37)	1253 ± 45	17	19.0
B	oxo polyoxo	40	40.1	8.3	32	47 (37)	1067 ± 30	17	20.0
						45 (38)	1203 ± 40	17	18.0

\*Native samples. Numbers in parentheses represent the unit cell parameters of the calcined samples.  $^a w = a_0 - \phi_{\text{BHH}}$ .

(see page 955).

Table 2 Cyclooctene epoxidation over various catalysts, after 24 hours reaction. In all cases there is 100% selectivity to epoxide\*

Run	$\text{H}_2\text{O}_2$ : Substrate (mol/mol)	Substrate: W (mol/mol)	Cyclooctene conversion (%)
All Si	5		0
A1c	5	63	98
A2c	5	42	84
Bc	5	62	33

\*Reaction conditions: see text. Room temperature experiments.

(see page 957). This is identical with the claims.

4. Claims 1-5, 7-10, 16, and 18-20 rejected under 35 U.S.C. 102(b) as being anticipated clearly by Jin et al (J. of Catalysis, 203, 2001, p75-81.).



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1. The  $\text{WO}_3/\text{SiO}_2$  catalyst is one of the powerful heterogeneous catalysts for the liquid phase cyclopentene oxidation by  $\text{H}_2\text{O}_2$ , which exhibits high activity and excellent selectivity to GA. The as-prepared catalyst seems more suitable for the industrial process than those homogeneous catalysts owing to the convenience in the separation of the catalyst from the reaction products, which makes it possible to use the catalyst repetitively and to regenerate the deactivated catalyst.

2. In the  $\text{WO}_3/\text{SiO}_2$  catalyst, the amorphous  $\text{WO}_3$  species are determined to be the active sites. The anhydrous  $\text{WO}_3$  exhibited almost no activity because of its good crystalline structure. The optimum  $\text{WO}_3$  loading is determined as 15 wt% to ensure the largest number of active sites but without significant crystallization. The optimum calcination temperature is determined as 823 K to guarantee the strongest interaction and support under the condition that no significant crystallization occurs, which can effectively inhibit the leaching of the active sites during the reaction. The t-BuOH is proved to be the best solvent in the present oxidation reaction owing to the promoting effect of TBHP formed through the reaction between t-BuOH and  $\text{H}_2\text{O}_2$  on  $\text{WO}_3/\text{SiO}_2$  as an acid catalyst.

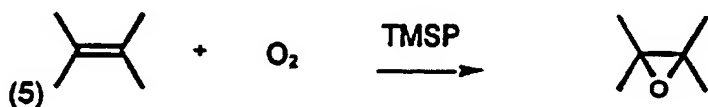
3. Under the present conditions, the 15 wt%  $\text{WO}_3/\text{SiO}_2$  catalyst can be used repetitively for three times (72 h). After reaction for 72 h, significant decrease in the activity of  $\text{WO}_3/\text{SiO}_2$  catalyst was observed, possibly due to the structural conversion of  $\text{WO}_3$  from the amorphous state to the crystalline state. The deactivated catalyst could be regenerated easily by calcining it at 823 K for 6 h.

(see page 80, left col.) . This is identical with the claims.

5. Claims 1-5, 7-10 , and 18-20 rejected under 35 U.S.C. 102(b) as being anticipated clearly by Neumann et al (WO 98/54165).

Neumann et al discloses the followings (see pages 6-7):

The process described in this invention relates to the use of transition metal substituted polyoxometalates (TMSP) to catalyze the epoxidation of alkenes with molecular oxygen according to the following equation (5).



Polyoxometalates are oligomeric oxides of defined structure based on addenda of tungsten, molybdenum, niobium or vanadium or a combination thereof. More specifically, transition metal substituted polyoxometalates are compounds of the general formula  $X_x(TM)_yM_mO_z^{q-}$  where the heteroatom, X, if present ( $x \geq 0$ ) may be main group or transition metals, the addenda atoms, M, are molybdenum, tungsten, niobium or vanadium or a combination thereof, and TM is one or several different transition metals. The specific class of transition metal substituted polyoxometalates,  $[WZnTM_2(XW_9O_{19})_2]^{9-}$ , used in the process described by the present invention are characterized as a dimer of a truncated Keggin structure having a "belt" of W,Zn and other transition metal (TM) cations "sandwiched" between the two  $B-XW_9O_{34}$  trivacant Keggin fragments the structure of which is shown in appended Figure 1. The transition metal cations are assumed to be positioned at terminal positions and are hexacoordinate with at least one labile ligand such as water. The TM atom can be any transition metal of the first, second or third row. More preferably the TM atom is a noble metal selected from the group

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consisting of ruthenium, rhodium, palladium, osmium, iridium and platinum. Most preferably the TM atom is ruthenium. The heteroatom atom, X, of the trivacant Keggin fragments can be any heteroatom known for Keggin compounds as is well-known in the art. For example, X can be a nonmetal such as phosphorous, silicon, germanium, boron, or arsenic. Alternatively X can be a metal such as zinc, cobalt, iron etc. The preferred transition metal substituted polyoxometalate for this process is  $[WZnRu_2(ZnW_9O_{19})_2]^{11-}$ . No transition metal substituted polyoxometalates or those of the general structure described and shown above have ever been used as catalysts for the epoxidation of alkenes with molecular oxygen. The counter cation of the above transition metal substituted polyoxometalates may be any cation including for example alkali metals, alkaline earth cations, transition metal cations or organic cations such as quaternary ammonium salts.

**Example 1**

A 5 ml solution of degassed 1,2-dichloroethane containing 100  $\mu$ mol  $Q_{11}WZnRu_2(ZnW_9O_{19})_2$  in which Q is tricaprylmethyl ammonium was kept under 1 atm molecular oxygen at 90 °C for nine hours in a closed vessel. The oxygen solution was cooled to room temperature and 11.2 mg cyclooctene was added.

After 2 hours the solution was analyzed by GLC. The analysis showed a 67.5 % conversion to cyclooctene oxide.

(see page 8, example 1). This is identical with the claims.

6. Claims 1-7, 9-10, 16-18, and 20 are rejected under 35 U.S.C. 102(b) as being anticipated clearly by Brown et al (WO 94/21583).

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Brown et al discloses the followings (see from page 3 ,line 18 to page 4, line 24):

According to the present invention, there is provided a process for the selective oxidation of alcohols wherein a substrate alcohol is contacted with hydrogen peroxide in a reaction medium in the presence of catalyst, characterised in that:

- i. the reaction medium comprises a solvent that does not comprise a carboxyl group, and
- ii. the catalyst comprises a heteropolyacid which is supported on an insoluble particulate support.

In the process according to the present invention, the catalyst contains a non-metallic heteroatom which may be selected from Group IV including silicon and germanium or Group V including phosphorus. Preferably, the catalyst contains phosphorus as the non-metallic heteroatom.

Tungsten or molybdenum, preferably tungsten, may constitute the entire metallic component of the heteropolyacid. Two transition metals, such as tungsten and molybdenum, may be incorporated therein. Many heteropolyacids for employment in the present invention process can be represented by the empirical formula  $Q_{3/n}PW_wMo_{12-w}O_{40}$  when they are brought into contact with the support in which  $w$  represents 0 or an integer of at least 1 and often at least 6. Most preferably,  $w$  represents 12.  $Q$  represents hydrogen or other counterion, and  $n$  is its basicity in the general formula. It is believed that the supported catalyst retains its empirical ratio of tungsten to phosphorous and molybdenum, but that the interaction of the catalyst with the surface of the support may result in the catalyst becoming bonded chemically to the support, thereby modifying both the catalyst itself and the support surface. Such treatments may also encourage a redistribution of the metal between species of different nuclearity.

Other heteropolyacids contemplated for use in the present invention include those containing at least one first series transition metal, including specifically iron, manganese, cobalt and nickel, for example in heteropolyacids of the formula  $Q_{(7-v)/n}PW_{11}MxO_{36+v}$  in which  $Mx$  represents the other transition metal,  $v$  is its oxidation state and  $Q$  is the counterion of basicity  $n$  as before.

The support for the heteropolyacid catalyst is often selected from solids of Group IIa, IIb, IIIb, IVa and IVb elements and from organic basic ion exchange resins. Within the class of Group IIa compounds, it is often convenient to select as support from magnesium compounds, including in particular, magnesium oxide and magnesium silicate. Within the class of group IIb compounds, it can be convenient to select from zinc compounds as support, including specifically zinc oxide. Within the class of Group IVa compounds, it is often convenient to select from titanium or zirconium compounds, including specifically titanium oxide and zirconium phosphate. From within the class of inorganic Group IVb compounds, it is often desirable to select as supports from tin compounds, many of which are readily available, or from germanium compounds. Specific examples include tin oxide.

During calcination of the supported catalyst material which has been obtained by impregnation of an inorganic support with the heteropolyacid, it is believed that formation of a bond between the catalyst and the support is promoted, which can assist in controlling the leaching of catalyst into the reaction mixture. However, as a result of such interaction and bond formation, the catalyst species may be altered to some extent by such calcination, so that the value of such a post impregnation calcination tends to vary depending upon the support employed. For some supports, including alumina in particular, it is advantageous to calcine at a temperature of at least 300°C and usually not higher than about 600°C. In a number of instances, a particularly suitable temperature for post-impregnation calcination of for example alumina is at least about 400°C and especially from about 450 to about 550°C. Other supports for which post impregnation calcination is an appropriate activity include magnesium silicate and zirconium phosphate. (see page 6 ,lines 22-35). This is identical with the claims.

7. Claims 1-7, 9-10 ,16-18, and 20 are rejected under 35

U.S.C. 102(b) as being anticipated clearly by Brown et al (WO 93/00338).

Brown et al discloses the followings (see an abstract page):

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Alkenes can be epoxidised with hydrogen peroxide using a homogeneous heavy metal catalyst, but discharge of spent reaction mixtures releases the heavy metal in the environment. The problem can be ameliorated by selecting a heterogeneous catalyst system comprising a tungsten-containing heteropolyacid supported on selected Group IIa, IIb, IIIb, IVa or IVb inorganic supports or on a strong basic resin, which catalyst has either been calcined after impregnation or in the impregnation stage an alcoholic solution of the heteropolyacid is employed and by employing a nitrile or oxygenated polar solvent reaction medium. A number of preferred heteropolyacids satisfy the empirical formula  $M_{3/n}PW_wMO_{12-w}O_{40}$  in which w represents an integer of at least 1, M represents a counterion and n its basicity. Preferred supports include activated alumina, calcined at 400 to 600° C and cross-linked quaternary ammonium-substituted polystyrene resins. The most preferred catalysts are made by impregnating an inorganic support with a methanol solution of the heteropolyacid to a desired loading of active material on the support and subsequently calcining the loaded support at 400 to 600° C. Preferred reaction media include acetonitrile and tertiary butanol.

#### Examples 1 to 8 and Comparisons C9 to C14

In these Examples and Comparisons, the selected catalyst (0.5g of as made catalyst/support), substrate (cyclohexene, 2g except in Ex 3, Ex4, Ex7 and C11, C15 which each employed 8g) and solvent (40ml) were charged into a three-necked glass reaction vessel equipped with a water-cooled condenser and the stirred mixture was heated to reflux temperature, which was about 80°C, except in Example 2, where it was about 60°C. Aqueous hydrogen peroxide solution (35% w/w except in Ex3 which used 70% w/w) was introduced slowly into the stirred refluxing reaction mixture over a period of 45 minutes to 1 hour. The mixture was refluxed for a further 5 hours. The reaction mixture was then analysed by gas liquid chromatography to determine how much substrate had been consumed during the reaction, including evaporation losses,

Table 1

Catalyst	Impregnation	Support	Prep	Calcine
Ref.	Species		Method	Temp °C
I	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	g-Al <sub>2</sub> O <sub>3</sub>	A	500
II	H <sub>3</sub> PW <sub>11</sub> MoO <sub>40</sub>	"	A	500
III	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	neutral Al <sub>2</sub> O <sub>3</sub>	A	500
IV	"	Mg silicate	A	500
V	(CP) <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	g-alumina	B	500
VI	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	g-Al <sub>2</sub> O <sub>3</sub>	A	600
VII	"	"	A	400
VIII	"	"	A	200
IX	"	"	A	-
X	"	"	A	500
XI	"	"	B	500

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XII	"	ZrPO <sub>4</sub>	A	500
XIII	"	SnO <sub>2</sub>	A	500
XIV	"	Al(OH) <sub>3</sub>	A	500
XVI	"	TiO <sub>2</sub>	A	500
XVII	H <sub>3</sub> PW <sub>6</sub> Mo <sub>6</sub> O <sub>40</sub>	α-alumina	A	500
R1	H <sub>3</sub> PMo <sub>12</sub> O <sub>40</sub>	SiO <sub>2</sub>	A	500
R2	H <sub>3</sub> PW <sub>12</sub> O <sub>40</sub>	acid-Al <sub>2</sub> O <sub>3</sub>	A	500
R3	"	basic-Al <sub>2</sub> O <sub>3</sub>	A	500
R4	"	α-alumina	A	500
R5	"	H-mordenite	A	500
R6	"	not supported		

(see from page 15 line 25 to page 16, line 36). This is identical with the claims.

### ***Claim Rejections - 35 USC § 103***

This application currently names joint inventors. In considering patentability of the claims under 35 U.S.C. 103(a), the examiner presumes that the subject matter of the various claims was commonly owned at the time any inventions covered therein were made absent any evidence to the contrary. Applicant is advised of the obligation under 37 CFR 1.56 to point out the inventor and invention dates of each claim that was not commonly owned at the time a later invention was made in order for the examiner to consider the applicability of 35 U.S.C. 103(c) and potential 35 U.S.C. 102(e), (f) or (g) prior art under 35 U.S.C. 103(a).

The following is a quotation of 35 U.S.C. 103(a) which forms the basis for all obviousness rejections set forth in this Office action:

(a) A patent may not be obtained though the invention is not identically disclosed or described as set forth in section 102 of this title, if the differences between the subject matter sought to

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be patented and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which said subject matter pertains. Patentability shall not be negated by the manner in which the invention was made.

The factual inquiries set forth in *Graham v. John Deere Co.*, 383 U.S. 1, 148 USPQ 459 (1966), that are applied for establishing a background for determining obviousness under 35 U.S.C. 103(a) are summarized as follows:

1. Determining the scope and contents of the prior art.
2. Ascertaining the differences between the prior art and the claims at issue.
3. Resolving the level of ordinary skill in the pertinent art.
4. Considering objective evidence present in the application indicating obviousness or nonobviousness.

8. Claims 1-10 , and 16-20 are rejected under 35 U.S.C. 103(a) as being unpatentable over Briot et al (J. Mater. Chem. 2000, 10 , p. 953-958) in view of Neumann et al (WO 98/54165).

Briot et al discloses the followings (see page 957) :



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The results of cyclooctene epoxidation over W-containing MCM-41 are listed in Table 2. Blank experiments with purely siliceous MCM-41 show that it is virtually inactive for this reaction at room temperature. In contrast, W-containing MCM-41 catalysts are very active, even at room temperature. Comparison of A1c and Bc indicates that materials synthesized by route A are distinctly more active. This can be explained by the presence of less active  $WO_3$  material in the Bc sample. More interesting is the result obtained with sample A2c. Despite the high tungsten loading in A2c ((Si:W)<sub>cap</sub> = 20:1) and the large amount of tungsten used during the catalysis test ((Substrate:W) = 42:1), cyclooctene conversion is less than that observed with A1c. This result can be understood by considering that sample A2c does not only contain isolated W species, but also tungsten oxide and/or HPA species which are poorer precatalysts for this reaction. Catalysis results match those obtained by different physicochemical techniques, especially Raman spectroscopy. Furthermore, atomic emission spectroscopic analysis of the epoxidation solutions indicates that there is reduced leaching (quantitative analysis is in progress) of W into the solutions. Work is in progress to obtain materials resistant to nucleophilic attack and to run tests with weak nucleophiles.

Table 1 Tungsten content, (Si/W) molar ratio, unit cell parameters, specific surface area, mean pore diameter ( $\phi_{BHH}$ ) and wall thickness ( $w$ )

Run	Route	(Si:W) <sub>initial</sub>	Si (wt.%)	W (wt.%)	(Si:W) <sub>exp</sub>	$a_0^a/\text{\AA}$	$S_{DET}/m^2 g^{-1}$	$\phi_{BHH}/\text{\AA}$	$w/\text{\AA}$
All Si	1	1	1	1	=	45.5 (36)	1384 ± 45	16	19.5
A1	oxo peroxo	40	39.4	8.2	31	46 (37)	1253 ± 45	17	19.0
A2	oxo peroxo	20	36.2	12.5	19	47 (37)	1067 ± 30	17	20.0
B	oxo polyoxo	40	40.1	8.3	32	45 (38)	1203 ± 40	17	18.0

\*Native samples. Numbers in parentheses represent the unit cell parameters of the calcined samples.  $a^a = a_0 - \phi_{BHH}$ .

(see page 955).

Table 2 Cyclooctene epoxidation over various catalysts, after 24 hours reaction. In all cases there is 100% selectivity to epoxide\*

Run	$H_2O_2$ : Substrate (mol/mol)	Substrate: W (mol/mol)	Cyclooctene conversion (%)
All Si	5	=	0
A1c	5	63	98
A2c	5	42	84
Bc	5	62	33

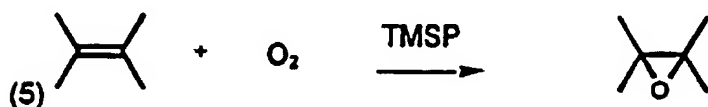
\*Reaction conditions: see text. Room temperature experiments.

(see page 957).

The instant invention, however, differs from the prior art in that the use of the hydrogen peroxide is not disclosed in the claimed process.

Neumann et al discloses the followings (see pages 6-7):

The process described in this invention relates to the use of transition metal substituted polyoxometalates (TMSP) to catalyze the epoxidation of alkenes with molecular oxygen according to the following equation (5).



Polyoxometalates are oligomeric oxides of defined structure based on addenda of tungsten, molybdenum, niobium or vanadium or a combination thereof. More specifically, transition metal substituted polyoxometalates are compounds of the general formula  $X_x(TM)_yM_mO_z^{q-}$  where the heteroatom, X, if present ( $x \geq 0$ ) may be main group or transition metals, the addenda atoms, M, are molybdenum, tungsten, niobium or vanadium or a combination thereof, and TM is one or several different transition metals. The specific class of transition metal substituted polyoxometalates,  $[WZnTM_2(XW_9O_{19})_2]^{q-}$ , used in the process described by the present invention are characterized as a dimer of a truncated Keggin structure having a "belt" of W,Zn and other transition metal (TM) cations "sandwiched" between the two  $B-XW_9O_{34}$  trivacant Keggin fragments the structure of which is shown in appended Figure 1. The transition metal cations are assumed to be positioned at terminal positions and are hexacoordinate with at least one labile ligand such as water. The TM atom can be any transition metal of the first, second or third row. More preferably the TM atom is a noble metal selected from the group

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consisting of ruthenium, rhodium, palladium, osmium, iridium and platinum. Most preferably the TM atom is ruthenium. The heteroatom atom, X, of the trivacant Keggin fragments can be any heteroatom known for Keggin compounds as is well-known in the art. For example, X can be a nonmetal such as phosphorous, silicon germanium, boron, or arsenic. Alternatively X can be a metal such as zinc, cobalt, iron etc. The preferred transition metal substituted polyoxometalate for this process is  $[WZnRu_2(ZnW_9O_{19})_2]^{11-}$ . No transition metal substituted polyoxometalates or those of the general structure described and shown above have ever been used as catalysts for the epoxidation of alkenes with molecular oxygen. The counter cation of the above transition metal substituted polyoxometalates may be any cation including for example alkali metals, alkaline earth cations, transition metal cations or organic cations such as quaternary ammonium salts.

**Example 1**

A 5 ml solution of degassed 1,2-dichloroethane containing 100  $\mu$ mol  $Q_{11}WZnRu_2(ZnW_9O_{19})_2$  in which Q is tricaprylmethyl ammonium was kept under 1 atm molecular oxygen at 90 °C for nine hours in a closed vessel. The oxygen solution was cooled to room temperature and 11.2 mg cyclooctene was added.

After 2 hours the solution was analyzed by GLC. The analysis showed a 67.5 % conversion to cyclooctene oxide.

(see page 8, example 1).

A further method to epoxidize alkenes is to use aqueous hydrogen peroxide as oxidant as shown in the reaction illustrated in equation (3).



Such a reaction represents a conceptual improvement compared to the use of organic hydroperoxides in that the co-product is water and therefore is environmentally benign and need not be recovered or recycled. As in the use of alkyl- and alkylaryl hydroperoxides the presence of a catalyst is necessary, which catalysts are again often compounds containing Ti(IV), V(V), Mo(VI) or W(VI), among others. In only certain cases has high selectivity been reported for alkene epoxidation. Some effective and selective catalysts include titanium silicalite-1 and other titanium substituted zeolites, and polyoxometalates such as  $[WZnMn_2(ZnW_9O_{19})_2]^{12-}$  and  $\{PO_4[WO(O_2)_2]_4\}^{3-}$ . In many cases, the use of hydrogen peroxide represents an ideal oxidant provided reactions are selective. An exception is in cases where the low price of the epoxide make the use of hydrogen peroxide prohibitively expensive.

(see from page 2, line 23, to page 3 ,line 14).

Briot et al teaches the epoxidation of the alkene using the catalyst containing tungsten, whereas Neumann et al discloses that it is possible in the presence of oxygen or hydrogen peroxide that the alkene can be epoxidated by using the catalyst containing tungsten. There is a teaching of an equivalence between oxygen and hydrogen peroxide for the epoxidation process.

Furthermore, Neumann et al has offered guidance that the use of the hydrogen peroxide in the process can be environmentally benign (see page 3 ,lines 4-5).

Therefore, it would have been obvious to the skilled artisan in the art to be motivated to incorporate Neumann's et al hydrogen peroxide into the Briot et al process in order to develop the process for the environmental safety. This is because the skilled artisan in the art would expect such a manipulation to be

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successful and efficient for the reason of the environmental safety as guidance shown in the Neumann et al.

Any inquiry concerning this communication or earlier communications from the examiner should be directed to Taylor Victor Oh whose telephone number is 571-272-0689. The examiner can normally be reached on 8:30-5:00.

If attempts to reach the examiner by telephone are unsuccessful, the examiner's supervisor, Cecilia Tsang can be reached on 571-272-0562. The fax phone number for the organization where this application or proceeding is assigned is 571-273-8300.

Information regarding the status of an application may be obtained from the Patent Application Information Retrieval (PAIR) system. Status information for published applications may be obtained from either Private PAIR or Public PAIR. Status information for unpublished applications is available through Private PAIR only. For more information about the PAIR system, see <http://pair-direct.uspto.gov>. Should you have questions on access to the Private PAIR system, contact the Electronic Business Center (EBC) at 866-217-9197 (toll-free).

*Taylor V Oh*  
*3/15/06*